Application No. 09/744,978

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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF	:
LAURA ZANIBELLI, ET AL.	: EXAMINER: NORTON
SERIAL NO: 09/744,978	:
FILED: APRIL 6, 2001	: GROUP ART UNIT: 1764
FOR: PROCESS AND CATALYSTS FOR UPGRADING OF HYDROCARBONS BOILING IN THE NAPHTHA RANGE	:

DECLARATION UNDER 37 C.F.R ∋ 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes Thierry CHOLLEY who states that:

1. I am a named inventor on the above-identified application.

2. I am a graduate of Doctor in chemistry at Paris VI university (France) and received my PHD degree in the year 1997.

3. I have been employed by TOTAL for 8 years as a research engineer.

4. I have read and understand the above-identified application.

5. The following experiments were performed by me or under my direct supervision.

6. The following experiments coupled with the data presented in the above-identified application demonstrate the importance of employing a catalyst with a FER zeolite in an amount of from 5 to 30% by weight with respect to the total weight of the catalyst.

7. This range of FER zeolite in the catalyst is not described nor was suggested to me from the description provided in U.S. 5,482,617; EP 0 665 280 A1; and 5,378,352.

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8. The following catalysts and the conditions under which they were tested are summarized in the following Table. The Table also provides the RON and MON results at the same desulfurization rate of residual sulfur in gasoline:

Catalyst	**Corresponding disclosure in the patent application**	**Test Conditions**	**Residual S**	**RON**	**MON**
55 % FER Mobil 303 m^2/g	Catalyst of Example 13 and Example 30	40 bars VVH 1 347 °C	63 ppm	82.4	84.5
10% FER Mobil 281 m^2/g	Catalyst of Example 14, and Example 31	10 bars VVH 4 308 °C	63 ppm	82.4	76.9
10% FER Sol-Gel 315 m^2/g	Catalyst of Example 12 and Example 35	10 bars VVH 4 330 °C	63 ppm	84.5	78.5
20 % FER + P Physical mixing 270 m^2/g	Catalyst of Example 17 and Example 34	10 bars VVH 1.5 320 °C	63 ppm	84	78.2

9. The results above demonstrate that the catalyst with 10 and 20 % FER zeolite had significantly higher octane numbers (RON and MON) compared to the catalyst with 55% (which is also the catalyst prepared and used in U.S. 5,378,352).

10. In a similar set of experiments, summarized in the Table below, also confirms the higher levels of octane numbers (RON and MON) at the same desulfurizing rate when catalysts with 10 % and 20 % FER zeolite were employed:

Catalyst	**Corresponding disclosure in the patent application**	**Test Conditions**	**Residual S**	**RON**	**MON**
10 % FER Sol-Gel 315 m^2/g	Catalyst of Example 12 and Example 35	10 bars VVH 4 324 °C	178 ppm	88.5	79.8
20 % FER Physical Mixing 302 m^2/g	Catalyst of Example 16 and Example 33	10 bars VVH 1.5 315 °C	178 ppm	87.7	79.2
20 % FER + P Physical Mixing 270 m^2/g	Catalyst of Example 17 and Example 34	10 bars VVH 1.5 307 °C	178 ppm	87.8	79.3

11. The catalysts were also compared based on the relative change of hydrocarbons sulfur content as a function of reaction temperature. The results are shown in Exhibit 1.

12. The results shown in the graph of Exhibit 1 demonstrate that when catalysts are tested under 10 bars, VVH 4, the 10% FER US Pt catalyst was the most active catalyst for desulfurization for a reaction temperature of 20 °C less.

13. When catalysts were tested under 10 bars, VVH 1.5 : 20%FER + P MP Catalyst was the most active (7°C less than 20% FER MP catalyst). Phosphorus improved the desulfurizing activity. However, both of these catalysts were less active in desulfurization than 10%FER US Pt., which desulfurized better even if the VVH is much higher, i.e., it treated 2.7 times more gasoline for the same duration. 55%FER US Pt (Mobil) was much less active even at high pressure, it slowed flow-rate, and desulfurized gasoline at much higher temperature.

14. Additional experiments were performed to assess the RON and MON values as a function of temperature for the catalysts described above (and in the above-identified patent

application). The results are presented in Exhibit 2 (RON variation) and Exhibit 3 (MON variation). The results demonstrate the improved maintenance of RON and MON at the highest VVH.

15. The specification also provides examples demonstrating the superiority of catalysts with 5 to 30 % FER zeolite. Example 30 is a comparative example which has 55% by weight of the FER component and is the catalyst described in U.S. 5,378,352. This comparative Example can be compared to Examples 31 (9.82 % FER), 33 (19.84 % FER), 34 (20.37 % FER), and 35 (10% FER).

16. The data obtained with the Comparative Example is summarized on page 47, lines 13-16, "The Octane losses obtained are very high in all the range of temperatures tested. RON loss is in the order 13 units and MON loss is in the order of 5 to 6 units."

17. In contrast, the data obtained from Example 31, (page 48), Example 33 (page 50), Example 34 (page 51) , and Example 35 (page 52), confirmed that the octane losses were significantly lower relative to Example 30.

18. In summary, desulfurizing activity alone is insufficient to determine the interest of a catalyst. The most important criteria is to obtain a desulfurized gasoline at a residual sulfur content for which the RON and MON are the highest utilizing the lowest hydrogen pressure (i.e., lowest hydrogen consumption) and the highest VVH as possible.
The catalysts prepared and employed by the present invention meet these criteria, whereas those known previously, as exemplified in U.S. 5,378,352, did not meet these criteria.

19. I declare further that all statements made herein of are of my own knowledge are true and that all statements made on information are believed to be true. Further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Thierry CHOLLEY

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November 14th 2003